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Uranium Measurements and Attributes

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Abstract

It may be necessary to find the means to determine unclassified attributes of uranium in nuclear weapons or their components for future transparency initiatives. We briefly describe the desired characteristics of attribute measurement systems for transparency. The determination of uranium attributes; in particular, by passive gamma-ray detection is a formidable challenge.

Introduction

Highly enriched uranium (HEU) is one of the two key fissile materials used in nuclear weapons since the dawn of the nuclear age. HEU is uranium that has been enriched to greater than 20% in its isotope ^{235}U. During the cold war, the United States, the Soviet Union, and others produced hundreds of metric tons of HEU. Because of its importance, some U.S. policymakers view the detection of HEU in nuclear weapons and their dismantled components as a potentially important transparency measure.

Radiation measurements in the context of arms control

Before discussing HEU detection and measurement it is useful to review the salient requirements placed on radiation measurements used for fissile material transparency.

1. Radiation measurements of nuclear weapons, their components, or their converted fissile material are intrusive. This is because they almost always acquire data that are classified.
2. Nuclear weapons and their components in storage containers are dense, inhomogeneous, and sizable. This situation contrasts with most traditional measurements of fissile material. With the exception of waste measurements, most fissile-material measurement techniques have been devised for inspection of fairly homogeneous items that are small compared to items of interest in arms control. For this reason, measurement results for arms-control applications generally should be independent of item configuration. By this, we mean that the measurement technology should be robust enough to obtain results for all types of items that it is intended to inspect. This is important for two reasons:
   - To ensure that the instrumentation can perform its intended function on all types of items.
   - If the instrumentation cannot perform its intended function on all types of items under inspection, it is possible, in some cases—from the physics principles upon which the inspection technique is based—to infer sensitive information about the classes of items for which it succeeds or fails.

   While there may be necessary and acceptable exceptions to this rule, they need to be examined carefully on a case-by-case basis.
3. Before a measurement technique can be accepted for use in an arms-control agreement, all parties must agree upon its use. Experience has shown that the likelihood of acceptability of a technology to an arms-control regime is increased if the following things are considered:

- Measurements cannot reveal classified information. Although the Atomic Energy Act was amended to allow limited sharing of classified information for arms-control purposes, as a practical matter this has not been a successful approach to date. Current efforts to protect classified information include making the radiation measurements behind information barriers.
- Simple technology generally is preferable to complex technology.
- Familiar technology generally is preferable to unfamiliar technology.
- Passive measurements generally are preferable to active-interrogation measurements.

Passive detection of shielded HEU

In this paper we focus on passive detection methods as they are the preferred approach for arms control. Passive radiation-detection methods exploit signatures that are intrinsic to the undisturbed material of interest. To protect classified information, measurements must be made externally to the weapon or material storage containers. Because nuclear weapons and components in their storage containers are large, dense, and inhomogeneous, the signature radiation must be sufficiently penetrating so that it can escape from the interior of the weapon or container and reach a detector. The radiation also must be of adequate intensity to allow completion of an inspection measurement in a reasonable period of time. For HEU, the only signature that can meet these criteria is from gamma rays emitted by the radioactive decay of uranium isotopes. Unfortunately the signature of ^{235}U is so weakly penetrating that the simple detection of shielded HEU—let alone its quantification—is a task that can range from fairly straight-forward to nearly impossible.

Even if ^{235}U is detectable, its presence alone is generally insufficient to determine that uranium is HEU. To make this determination, one must know the uranium enrichment. For HEU, the other dominant uranium isotope of interest is ^{238}U. Knowledge of the concentration ^{235}U and ^{238}U can provide an approximate estimate of uranium enrichment. However, even if the ^{235}U is detectable, the gamma rays from these two isotopes are sufficiently well separated in energy (notably at 186 keV for ^{235}U and 1001 keV for ^{238}U, see Fig. 1) that unknown differential attenuation precludes knowledge of their true relative emission intensities. An exception to this statement is the "enrichment meter" method that examines the 186-keV peak and the adjacent continuum to determine uranium enrichment. This method requires calibration against appropriate known standards, a condition unlikely to occur in many arms control scenarios. Because of its low energy, methods that exploit the 186-keV gamma ray generally are not applicable to detection of shielded HEU because they may be dependent on the item configuration.

Inference of the presence of HEU by use of a surrogate

An indirect alternative signature is being explored because determination of the presence of shielded HEU by measurement of its key isotopes often would be intractable in an arms-control setting (i.e. the detection of the presence of the impurity isotope ^{232}U). With only a 69-year half-life, ^{232}U does not occur in nature but is introduced as a result of reactor irradiation of uranium resulting in a number of complex reaction and decay chains. Four of the most significant of these complex chains[1] are:

(1) $^{235}U(\alpha)\ ^{231}Th(\beta^-)\ ^{231}Pa(n,\gamma)\ ^{232}Pa(\beta^-)\ ^{232}U$

(2) $^{234}U(\alpha)\ ^{230}Th(n,\gamma)\ ^{231}Th(\beta^-)\ ^{231}Pa(n,\gamma)\ ^{232}U$

(3) $^{235}U(n,\gamma)\ ^{236}U\ (n,\gamma)\ ^{237}U(\beta^-)\ ^{237}Np(n,2n)\ ^{236m}Np(\beta^-)\ ^{236}Pu(\alpha)\ ^{232}U$

(4) $^{238}U(n,2n)\ ^{237}U(\beta^-)\ ^{237}Np(n,2n)\ ^{236m}Np(\beta^-)\ ^{236}Pu(\alpha)\ ^{232}U$

Because uranium may contain ^{236}U as a result of the inclusion of previously reactor-processed uranium, another reaction chain may also be important:

(5) $^{236}U(n,2n)\ ^{237}U(\beta^-)\ ^{237}Np(n,2n)\ ^{236m}Np(\beta^-)\ ^{236}Pu(\alpha)\ ^{232}U$

In the 1960's, uranium from spent reactor fuel was reintroduced into U.S. gaseous diffusion plants to be re-enriched. Consequently, trace quantities of ^{232}U were entrained in the gaseous diffusion cascades where they remain today and have become a minor contaminant to new feed stock as it is introduced to the cascade. It is believed that similar circumstances have occurred elsewhere. Evidence[2] also suggests that, during the enrichment process, the ^{232}U is preferentially swept into the light isotope fraction that becomes HEU and unmeasurably small amounts get into the heavy isotope fraction that becomes depleted uranium. Therefore, the presence of ^{232}U in uranium is consistent with that uranium being HEU.

Uranium-232 decays by α-particle emission to become 1.9-year ^{228}Th where it enters the thorium decay series. The ^{228}Th rapidly decays through a succession of short-lived daughters, culminating at a final radioactive daughter, ^{208}Tl (see Fig. 1).

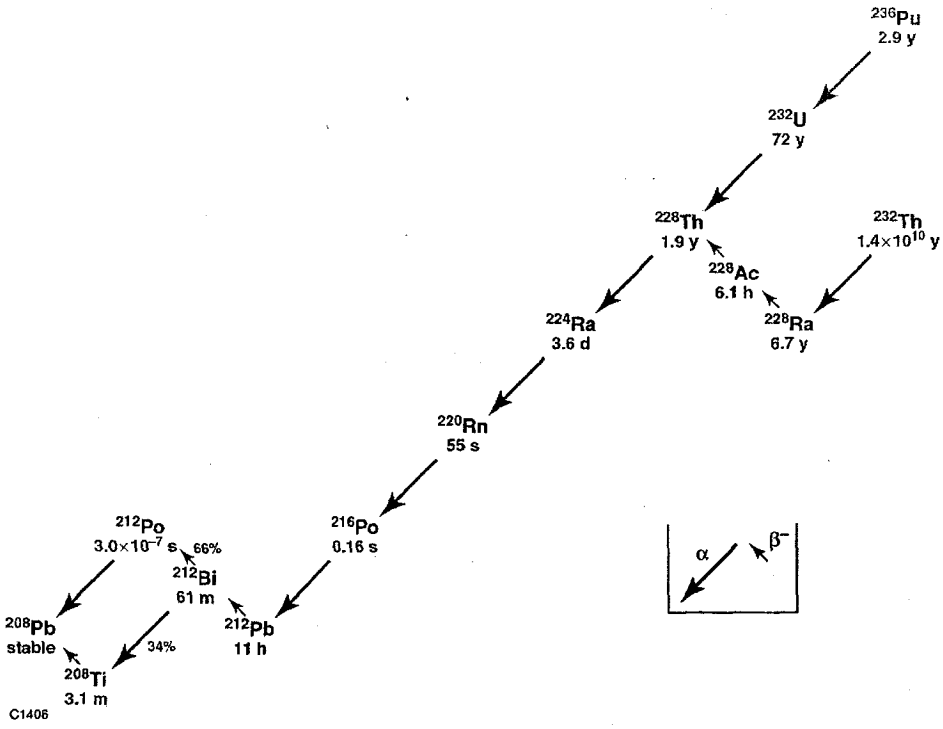

Figure. 1. Extended thorium decay series including ^{232}U and ^{236}Pu. Alpha decay is indicated by long arrows pointing to the lower left and beta decay by short arrows to the upper left.

There are a number of emissions associated with this decay series but the most distinctive is a highly penetrating 2615-keV gamma ray emitted by the β-decay of ^{208}Tl. As previously mentioned, ^{232}U is a trace contaminant in U.S. HEU and typically is found at the 100–200 parts per trillion (ppt) levels. Nevertheless, because of its relatively short half-life and the short half-lives of its daughters, it has relatively high specific activity thus making it readily observable in a gamma-ray spectrum. Figure 2 shows that at the 100-ppt level, the 2615-keV peak is of comparable height to the 1001-keV peak from ^{238}U in 93% enriched uranium in a thick sample.

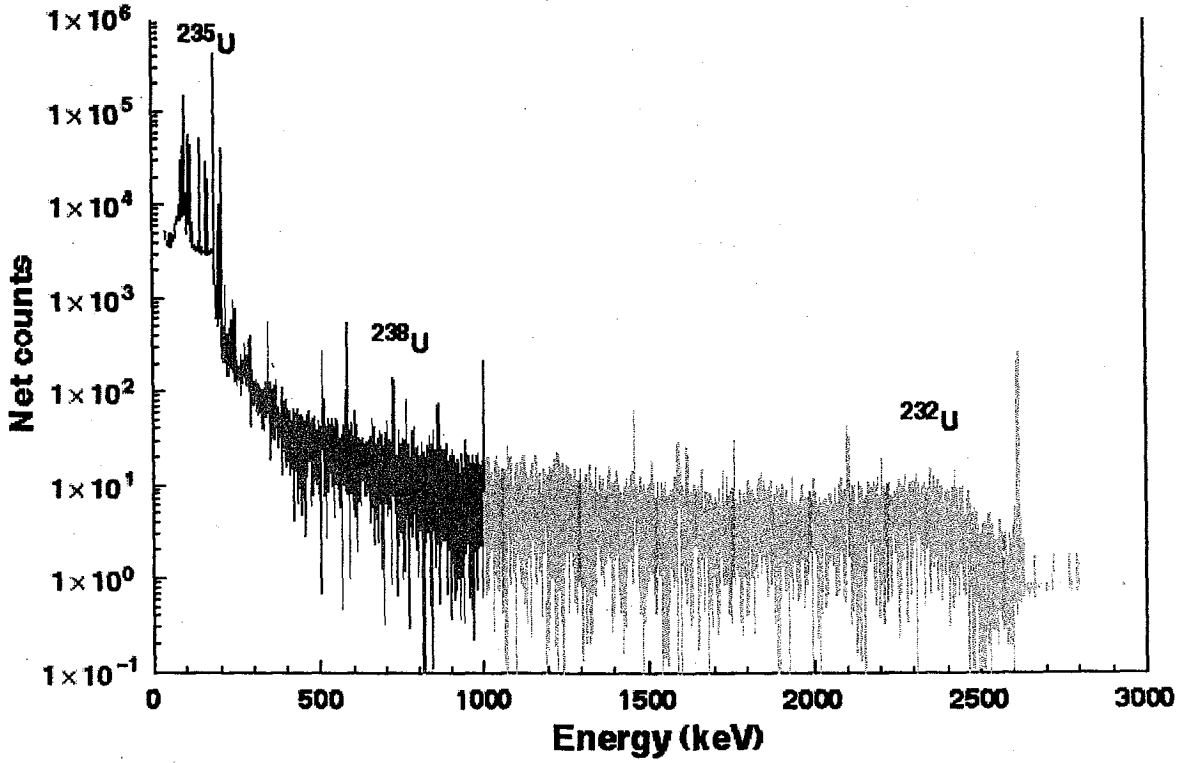

Figure 2. Background-subtracted, high-resolution gamma-ray spectrum from an unclassified 2.2-kg spherical source of uranium enriched to 93% in ^{235}U. The ^{232}U content is 100 ppt. The darker to lighter shadings of the plot indicate where the most prominent peaks are located for ^{235}U, ^{238}U, and ^{232}U.

Two difficulties must be overcome determining the presence of HEU through the detection of ^{232}U. (1) The gamma-ray emissions from ^{232}U are relatively weak, probably requiring longer measurement times than normally would be desirable. Unless a more satisfactory means of detecting shielded HEU is found, arms control regimes requiring the detection of HEU must be crafted to account for this difficulty. (2) The salient features of the ^{232}U signature, notably those associated with the decay of ^{208}Tl and its 2615-keV gamma ray, are not unique to ^{232}U.

The signature associated with the decay of ^{228}Th and all of its daughters, including ^{208}Tl, is found in natural background radiation because of trace quantities of thorium found ubiquitously in the earth's crust. Thus measurement times must be long enough to distinguish background thorium emissions from those in the items under inspection.

Another source of this signature is weapons-grade plutonium. During the creation of plutonium, a trace quantity of the impurity isotope ^{236}Pu is produced. Plutonium-236, with a half-life of 2.9 years, decays by α-emission to ^{232}U (Fig. 1.) and remains in the plutonium. This possibility must be considered for arms control regimes; however, it may be of small consequence in some situations because, in this case, the presence of the ^{236}Pu signature is evidence of the presence of a fissile material. Another concern is that since both natural thorium and depleted uranium are both plentiful and relatively inexpensive they could be placed in the sealed container to spoof an arms control measurement. The naturally occurring thorium chain begins with 1.4×10^{10}-year ^{232}Th which decays to ^{228}Ra then ^{228}Ac before reaching ^{228}Th. A telltale clue that natural thorium is present is a cluster of gamma rays emitted by ^{228}Ac in the neighborhood of 900 keV, with a 911-keV line being the most intense.

Conclusion

The simplest measurement technique for detection of shielded HEU would require detection of the penetrating 1001-keV line from ^{238}U to confirm the presence of uranium (but not HEU), the 2615-keV line that is consistent with the presence of HEU (or possibly weapons-grade plutonium), and the absence of a line at 911 keV to assure that the 2615 keV line is associated with fissile material. Although it is less penetrating, the absence of a gamma ray at 414 keV would build confidence that the 2615-keV line is associated with HEU rather than with weapons-grade plutonium. Sufficient time must be allowed for the measurement to accomplish these objectives; however the times can be reduced by careful construction of measurement systems that would need to include large high-resolution gamma-ray detectors.

References

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